Effect of Filler on the Aging Potential of Asphalt Mixtures

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In this work, the effect that filler has on the aging of bitumen was analyzed but with the filler being incorporated by volume, not by weight. The Universal de Caracterización de Ligantes method was used as the process of accelerated aging, and a new direct tensile test was used to determine the toughness of the aged mixture and, thus, to assess the effect the filler has. All tests performed for this paper have shown the protective effect of the fillers used. The new direct tensile test developed by the Road Research Laboratory of the Technical University of Catalonia allows observation of how an increase in filler produces an increase in the breaking load and a decrease in the maximum deformation: the hydrated lime tends to stiffen the mixture less and make it less brittle than does calcium carbonate. To minimize the effect of aging on bitumen, the filler content proposed must be 20% or 30% less than the content recommended in conditions where there is no aging, so that when the mixture ages, the mastic is able to build up the maximum energy possible.

The processes that a binder undergoes when aging, which cause its progressive hardening, may damage its aggregate–bitumen adhesive qualities and, thus, the cohesion of the mixture.

The addition of filler to the mixture can improve adhesion and cohesion substantially (filler is a fine material—it passes a 0.063-mm sieve—derived from aggregate or other similar granular material). The bitumen–filler system (mastic), which is thicker and tougher than bitumen alone, improves the adhesive qualities and, in providing a covering film of greater thickness, also can slow down the aging processes. The effects of the addition of filler are directly related to their characteristics and the degree of concentration of the filler in the bitumen–filler system.

The advantages that filler offers for the durability of the bituminous mixtures in the case of water action are due, in principle, to its physical characteristics, which reduce the porosity of the granular structure and thereby make access by water and air difficult. Moreover, the chemical nature of filler may mean greater affinity with the asphalt binder, which can improve resistance to the displacement that water causes the bitumen.

Using immersion tests, Craus et al. assessed the influence that the type of filler had on the durability of the bituminous mixtures (*1*). They reviewed the usual criteria of mixture design, with analyses that simulated short periods of exposure to the environment (for example, for the case under study, the residual Marshall stability and the resistance of immersion–compression); they noted that mixtures that pass these tests usually fail completely in service. With the obtained results, they were able to modify the existing criteria for the classification of fillers, which had been based only on basic properties without consideration of the durability factor. From this work, the authors have continued studying the effect of the characteristics of fillers on the durability of the mixtures (*2*).

Diverse methods of simulation of artificial aging have been developed to assess the durability of bituminous materials and mixtures. There are some well-known tests that are performed on the binder, such as the thin-film oven test (TFOT) and the rolling TFOT (RTFOT), which attempt to reproduce short-term aging, whereas others try to do it for the long term, like the pressure-aging vessel (PAV) test.

There are also diverse studies that use these procedures to assess the effect of filler in the aging of the binder. Thus, Petersen et al., using as a tool a variant of the RTFOT named thin film accelerated aging test, tried to quantify how the addition of filler might benefit the reduction of hardening by age and improve the properties of flow at low temperatures (*3*). They studied the physical and chemical behavior of mastics made with a conventional binder and three types of mineral fillings (lime with high calcium content, dolomitic lime, and limestone dust). First, they determined that the ideal filler–bitumen ratio by weight for the calcium lime was on the order of 20%; then, they compared the behavior of the three fillers at this proportion. The conclusions must be confined to that limited number of variables, but they established some comparative criteria according to which greater advantages were obtained with the use of calcium lime, whereas the use of limestone dust contributed little in resistance to aging.

While trying to adapt the PAV test to age mastics, Gubler et al. in the Swiss Federal Laboratories arrived at a better understanding of how the addition of filler can delay the aging of binders: the particles of mineral filling are an obstacle to the diffusion of oxygen in the heart of the bitumen (*4*). A numerical model determined that about 55 g of mastic was needed to obtain the same thickness of mastic film as of bitumen in the standardized method (which calls for the use of 50 g of bitumen), due to the greater density of mastic. However, it was observed that the obstacle effect of the filler could compensate for the fact that the thickness of film is less if the specified 50 g are used.

Nevertheless, a conceptual element was missing: the behavior of different fillers was analyzed when they were added to the weight, but the unequal filling effect that each of them had was ignored, a situation that became clear when comparisons were made of volumes.

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In 1960, Ruiz had already demonstrated the need to maintain the viscous properties of the bitumen filler system to guarantee that deformation does not occur with traffic and climatologic agents (*5*). Dorfman and Rivara established the limit known as critical concentration (*6*). This is a volumetric concentration of filler in the bitumen–filler system from which increasingly greater dispersions of viscous fluid appear. An excess of filler leads to greater mixture stiffness and a loss of adhesive qualities.

In addition to methods of aging for bitumens, there are also methods of accelerated aging, which are applied directly to the bituminous mixture. Among these are short-term oven aging (STOA) and long-term oven aging (LTOA). In addition to these, Lee et al. compared different methods of aging on the basis of the response of the binders (*7*). To synthesize the obtained conclusions, STOA is a more rigorous process than RTFOT for the short term, while the PAV test is a more rigorous process than LTOA for the long term.

The Universal de Caracterización de Ligantes (UCL) method belongs to this latter group of aging methods and was developed by Pérez Jiménez and Miró Recasens (*8*); the Cántabro test is used to measure cohesion that the binders give a standard mixture without fines. The method allows assessment of the variation in this cohesion with temperature, the action of water, and aging. The last part of the method uses an aging process on a compacted mixture, like the LTOA, the difference being the mixture used: a standard mixture, without fines, of high porosity (27% ± 1%) that is heated in an oven at 80°C for different periods of time. The high porosity of the mixture allows the bitumen to age in the thin film but in contact with the aggregate; this condition allows assessment of the effect of aging directly on the mixture without the need to extract the bitumen from the mixture (*9, 10*).

The aim of the work presented here, as with some of the earlier-mentioned referenced work, is to analyze the effect that filler has on the aging of bitumen but with incorporation of the filler by volume, not by weight. The UCL method is used as the process of accelerated aging and a new direct tensile test is used to determine the toughness of the aged mixture and, thus, to assess the effect the filler has.

METHODOLOGY

To assess the effect the aging of the bitumen–filler system (mastic) has on the toughness that it gives to the standard mixture, a new direct tensile test has been selected, the Barcelona Tracción Directa (BTD) test, developed by Pérez Jiménez et al. (*11, 12*) in the Road Research Laboratory of the Technical University of Catalonia, Spain.

The test consists of making a specimen in the Marshall molds on a metal base made of two semicircles with a projection at the contact area, which creates a cleft in the central part of the specimen, and an anchoring system, compacted only on the upper surface (50 blows). The quantity of mixture needed is such that the height of the specimen above the base section is about 3 cm. When the specimen is removed from the mold, the notched base remains attached to the lower part; this means that clamps can be attached, so that force can be applied for a tensile test at a constant speed of deformation. During the test, the opening of the cleft occurs, which causes the specimen to crack, as shown in Figures 1 and 2. The values registered are tensile effort applied and the size of the opening of the cleft.

To make the UCL standard mixture, granite aggregate was used with a special grading (Table 1). The bitumen used was a conventional-penetration bitumen, 80 to 100 pen; its concentration was kept constant at 4.5% by mass of aggregate.

FIGURE 1 BTD test: specimen notched with special bases.

For the mastics, two with significantly different characteristics were chosen: calcium carbonate and hydrated lime. First, their critical concentrations were determined according to the standardized procedure.

The critical concentration corresponds to a dispersion of filler particles in the bitumen moving as freely as possible but in contact with each other, that is, when the applied stress is consumed in the viscous deformation of the continuous bitumen medium and the frictional resistance between particles is at a minimum. This particle arrangement is what is expected in the sediment and is obtained by settling

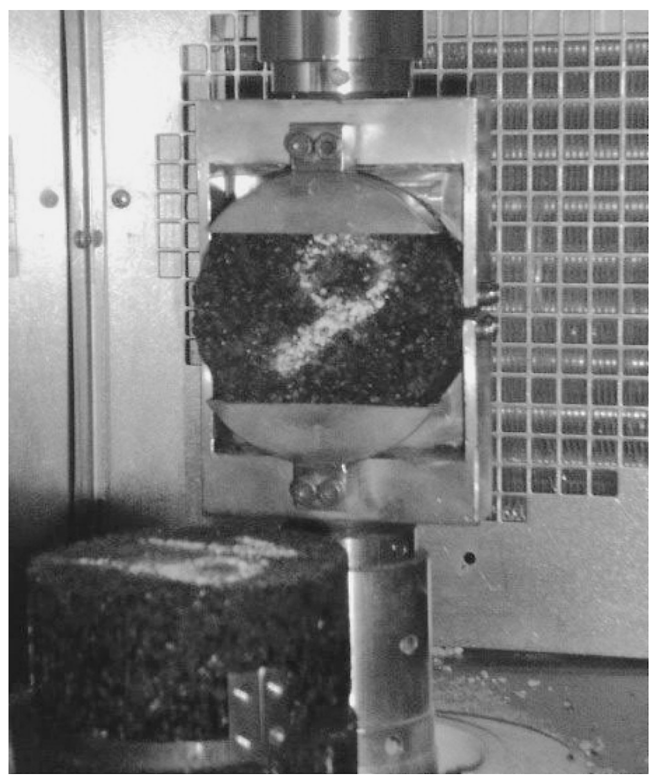

FIGURE 2 BTD test: testing of specimen.

TABLE 1 Standard UCL Mixture Design

Grading Sieve (mm)	Passing (%)	Bitumen Content (% of Aggregate)	Air Voids Content (%)
5	100		
2.5	20	4.5	27 ± 1
0.63	0		

a dispersion of filler in a fluid liquid medium with a chemical relationship with bitumens such as kerosene. The critical concentration is calculated with Equation 1:

$$C_s = \frac{P}{V \cdot \gamma} \tag{1}$$

where

C_s = critical concentration,
P = dry weight of filler (g),
V = settled volume of filler in anhydrous kerosene after 24 h (cm^3), and
γ = specific gravity of filler (g/cm^3).

The specific gravity of the hydrated lime is 2.351 g/cm^3, and its critical concentration is 0.165; the specific gravity of calcium carbonate is 2.771 g/cm^3, and its critical concentration is 0.370.

Second, these fillers were incorporated into the standard UCL mixture, in increasing volumetric concentrations (C_v), with C_v/C_s ratios of 0, 0.5, 1.0, 1.3, and 1.5, where $C_v/C_s = 0$ is the corresponding ratio to the reference mixture, without filler. The amounts added in weight are noticeably different for each one of the fillers (Table 1), which is why analyzing the same filler–bitumen ratio in weight (as recommended by the Spanish specifications, for example) would not make sense. Therefore, the addition of filler was made by volume, not by weight.

Once the specimens of the mixture were made on the notched bases, they were aged in an oven at 80°C for 0, 2, 4, and 7 days, the Day 0 specimen being without accelerated aging and undergoing only the aging caused by the manufacturing process.

The BTD direct tensile test was performed at 20°C and at a speed of deformation of 1 mm/min, slow enough to show the more-or-less ductile fracture of the mastics. The analyzed parameters were the critical load, the maximum deformation (obtained by means of the tangent straight line in the postbreaking zone of the curve load versus deformation) and the specific energy of fracture, obtained as the area of the load displacement curve divided by the area of fracture of the specimen.

In addition, in the case of the lime, once the specimens were tested, the bitumen was then extracted. This was done so that its characteristics could be analyzed and compared with the original characteristics to assess, in a more conventional way, the effect of this filler on the aging of the bitumen itself.

The bitumen extraction is made by the centrifuge method and uses dichloromethane (ASTM D2172-95, "A" method) at ambient temperature. Then, a distillation using the Rotovapor apparatus (ASTM D5404-93) is performed. The use of dichloromethane does not require an unusually high initial temperature (with dichloromethane, it takes only 40°C, while with trichloroethylene, it takes 89°C), and the bitumen remains at 163°C only for the last 20 min at a controlled pressure. In this way, the extraction and distillation processes do not affect the bitumen.

ANALYSIS OF RESULTS

Figures 3 through 6 show the development of the penetration, the softening point, and the viscosity of the recovered bitumen of the standard

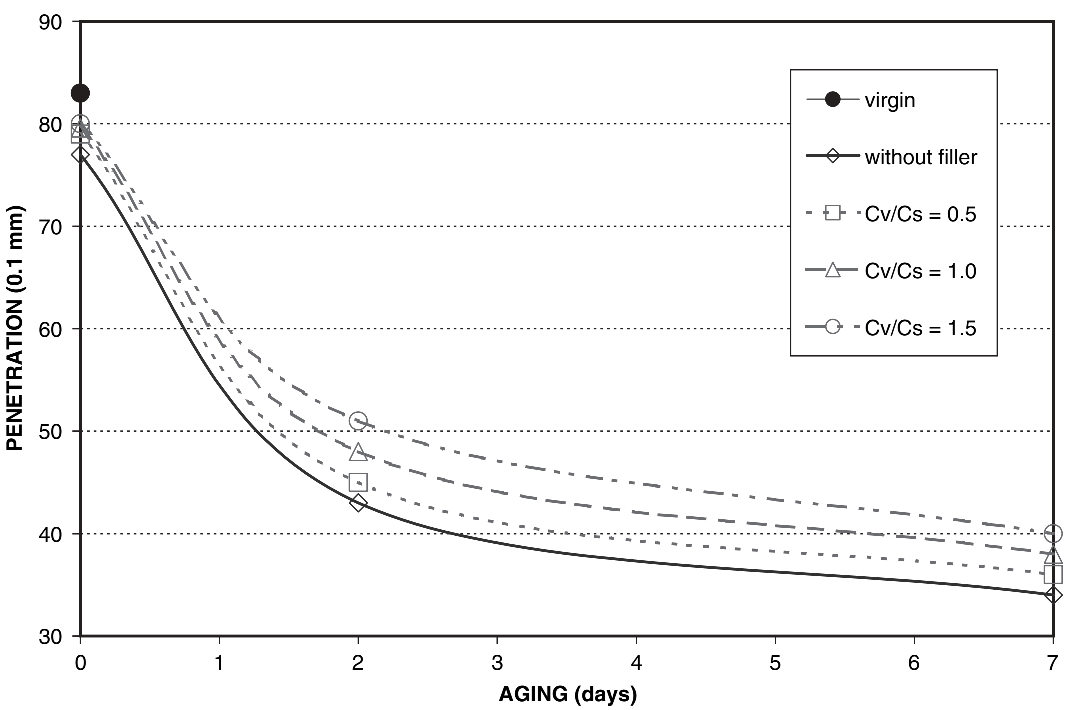

FIGURE 3 Penetration of bitumen recovered from lime mixture with different periods of aging and different filler contents.

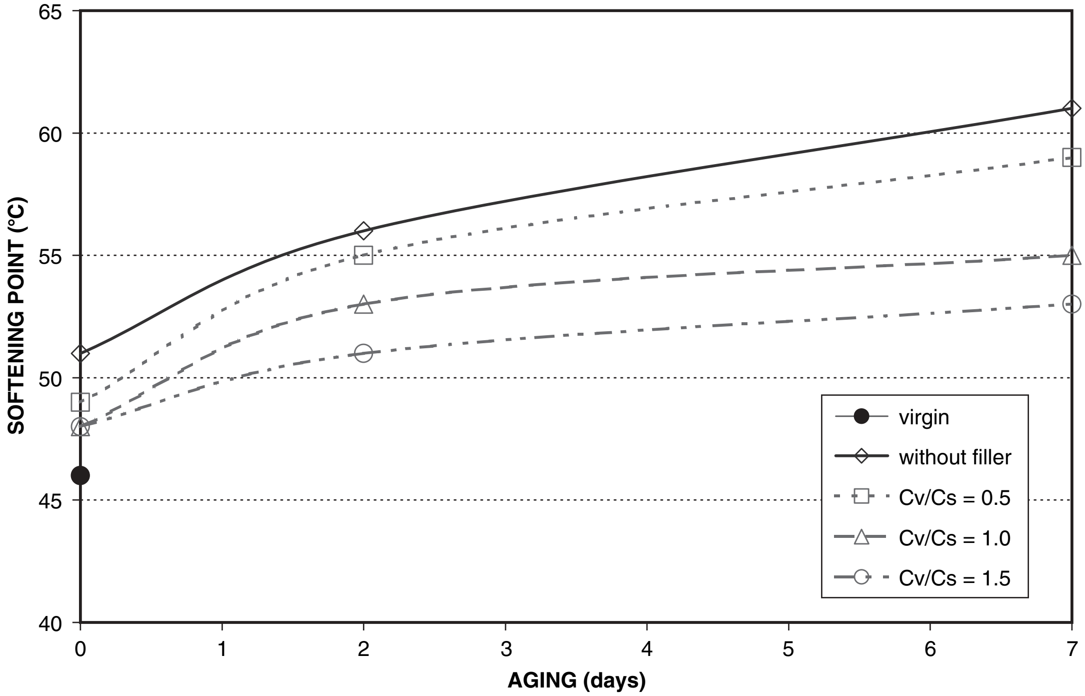

FIGURE 4 Softening point of bitumen recovered from lime mixture with different periods of aging and different filler contents.

mixture, which was aged over different periods of time. Moreover, it can be observed how the filler, in this case the hydrated lime, produces a smaller decrease in the penetration, a smaller increase in the softening point, and a smaller increase of viscosity in comparison with these variables in the bitumen of the reference mixture without filler.

These results demonstrate that the greater the amount of filler added, the less the bitumen ages. On one hand, this highlights the role of protector that the filler has in the aging of the bitumen, but, on the other hand, it means the durability of the mixture cannot be assessed, since it is evident that high contents of filler mean a brittle mixture that can quickly deteriorate. This further means that it is

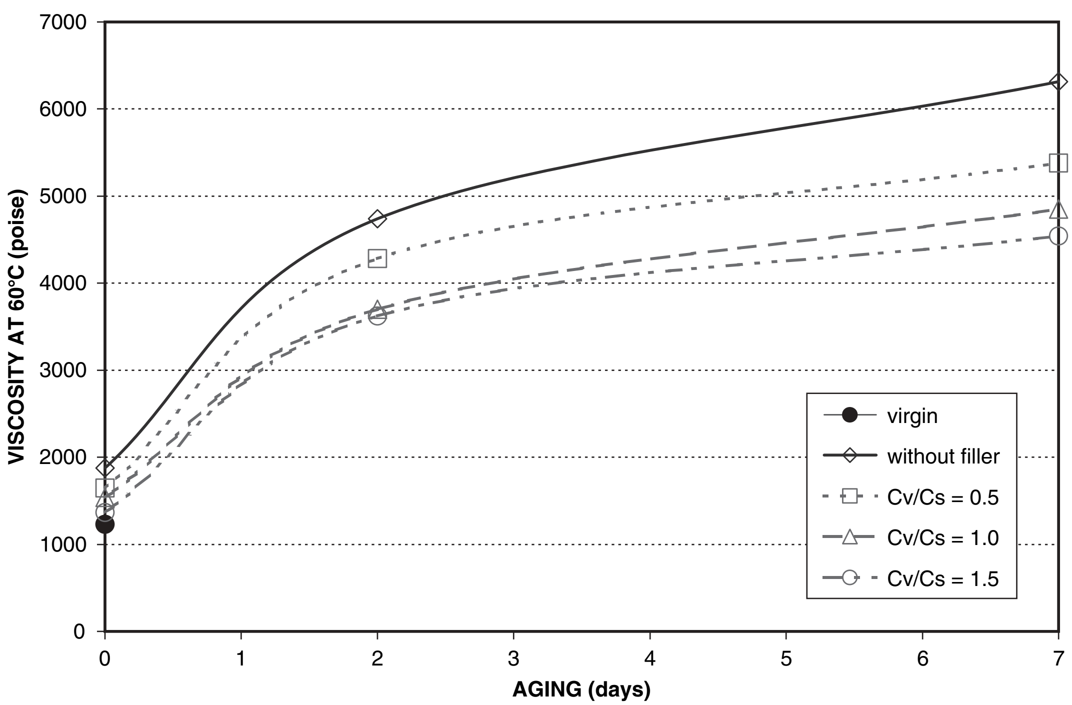

FIGURE 5 Dynamic viscosity at 60°C of bitumen recovered from lime mixture with different periods of aging and different filler contents.

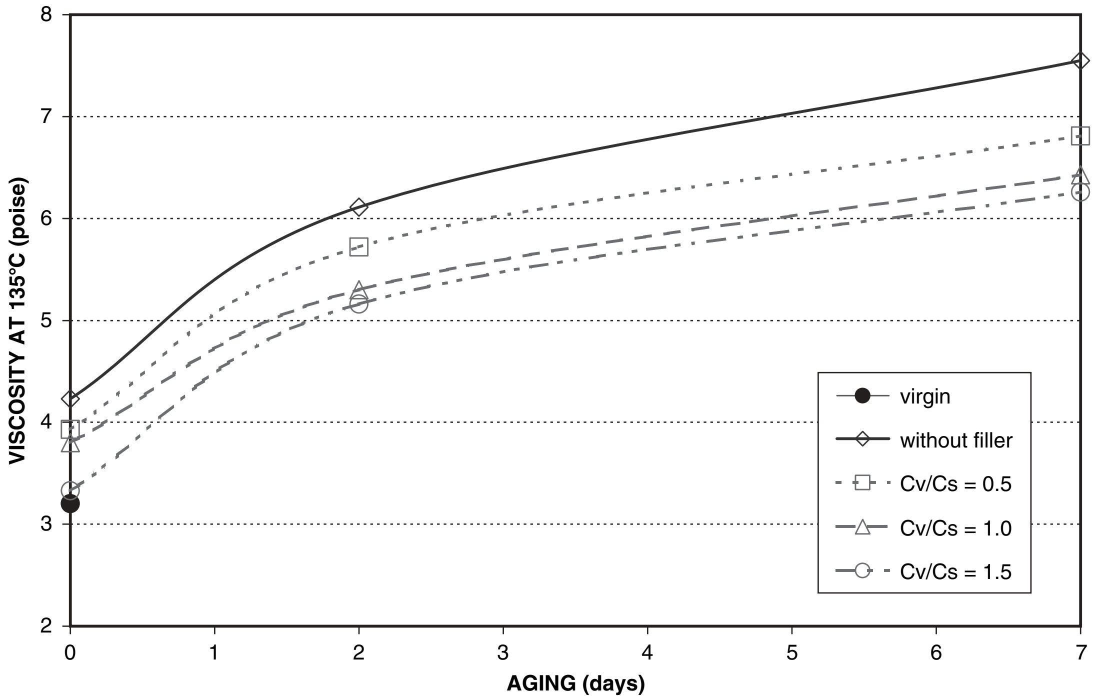

FIGURE 6 Dynamic viscosity at 135°C of bitumen recovered from lime mixture with different periods of aging and different filler contents.

not possible to establish a criterion for selecting the most suitable content of filler in terms of durability of the mixture. This indeed is the aim of the analysis of the effect of aging on the toughness of the mastic using the BTD test.

Figure 7 shows as an example the load displacement curves obtained as the average for three specimens for the reference mixture without filler and for different periods of aging. This figure shows how the breaking load tends to increase and the maximum deformation tends to decrease, clearly highlighting how the aging of the binder tends to stiffen the mixture, while at the same time the aging of the binder makes the mixture increasingly brittle.

In Figures 8 and 9, breaking load and maximum deformation are shown, for each of the fillers analyzed, on the basis of different volumetric concentrations at which the fillers were added to the standard mixture. To simplify the figures, only the values corresponding to the mixture aged for 7 days and the reference mixture, which was not aged (to 0 day), are represented. These values have been obtained, generally, from a mean of three individual results, and the variation coefficients range between 3% and 12% for the breaking load and 5% and 18% for the maximum deformation.

For calcium carbonate, the breaking load tends to increase as the volumetric concentration increases until it reaches the value of 1.3 times the critical concentration, at which point the load no longer increases. In contrast, the maximum deformation tends to diminish from $C_v/C_s = 0.5$.

Although this happens as much for the mixture that has not been aged as for the aged one (the curves are quite similar), the increase in the load and the decrease in the deformation are more marked for the mixture that has not been aged, which once again highlights the protective effect the filler has when bitumen ages. It is important to consider that the addition of filler causes a decrease in the voids in the mixture, from 28% to 21%, when the content is raised from 0 to 1.5 times the critical concentration (Table 2).

The same tendencies as those noted for calcium carbonate are observed for the hydrated lime. However, neither the increase in the breaking load nor the decrease in the maximum deformation when the volumetric concentration is increased is as pronounced for the hydrated lime as it is for the calcium carbonate, for both the aged and the nonaged mixtures. This could indicate that the protective effect of the hydrated lime against aging can be greater than that provided by calcium carbonate, especially if taken into account is the fact that the porosity of the mixture is somewhat greater when hydrated lime is used, with a percentage of voids that only falls 2% when it goes from 28% of the mixture without filler to 26% of the mixture with a hydrated lime content of 1.5 times its critical concentration (Table 3).

Nevertheless, with these figures, it is difficult to establish a criterion for selecting the most suitable filler content in terms of the durability of the mixture. For this, Figure 10 shows the variation in the specific energy of fracture with the volumetric concentration of incorporated filler for each of the fillers used.

For the calcium carbonate, with the mixture that has not been aged, we can see how the specific energy of fracture presents a maximum at $C_v/C_s = 1.3$. However, the C_v/C_s ratio for which the aged mixture is able to develop greater energy is slightly inferior, about 1.0. This means that, although the addition of filler (in this case calcium carbonate) protects the bitumen from aging, the amount to be added should be slightly less than the recommended ratio under conditions of no aging. When the C_v/C_s ratio goes from 1.3 to 1.0, it

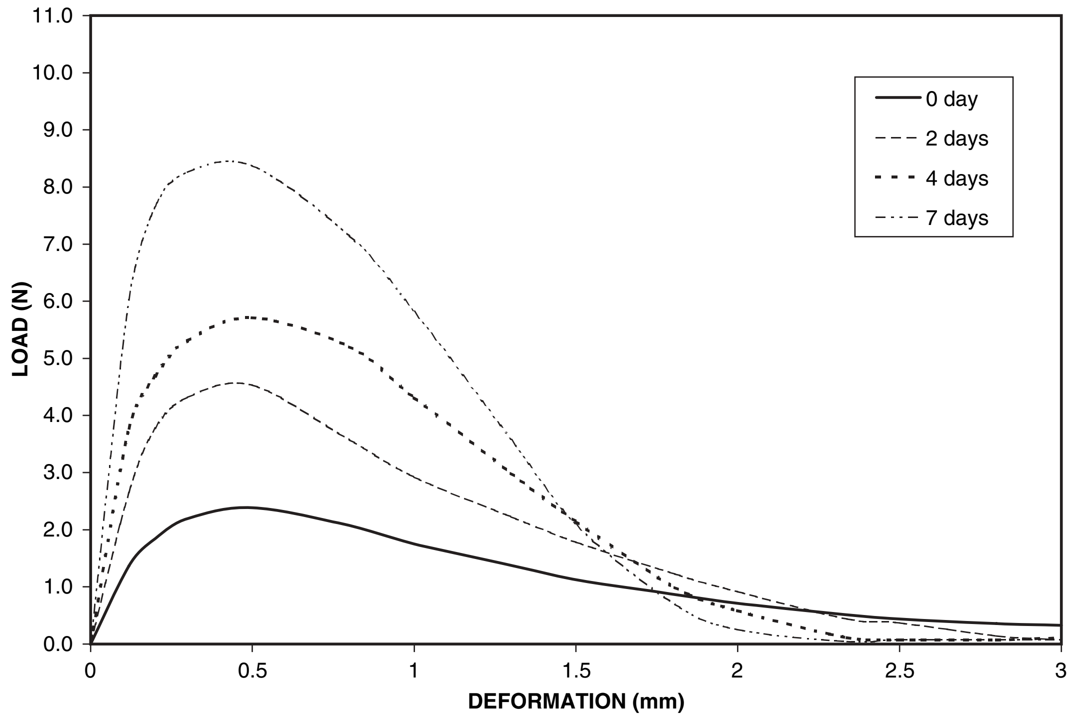

FIGURE 7 Load displacement curves by BTD test for different periods of aging (mixture without filler).

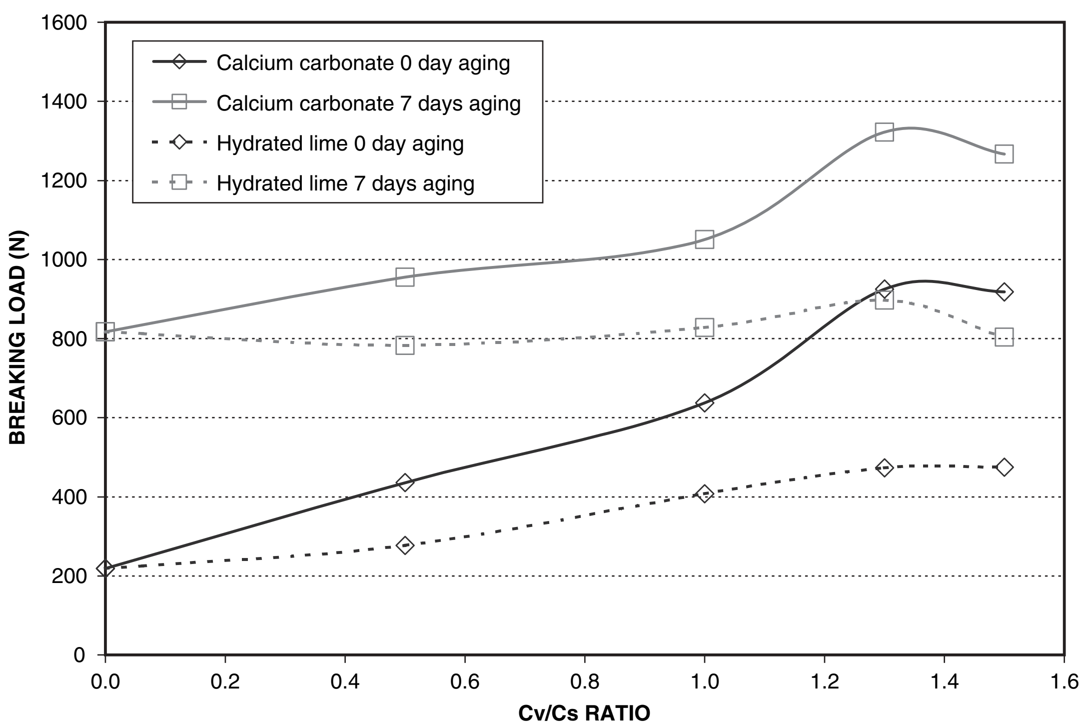

FIGURE 8 Variation of breaking load with volumetric concentration of filler for 0 and 7 days of aging.

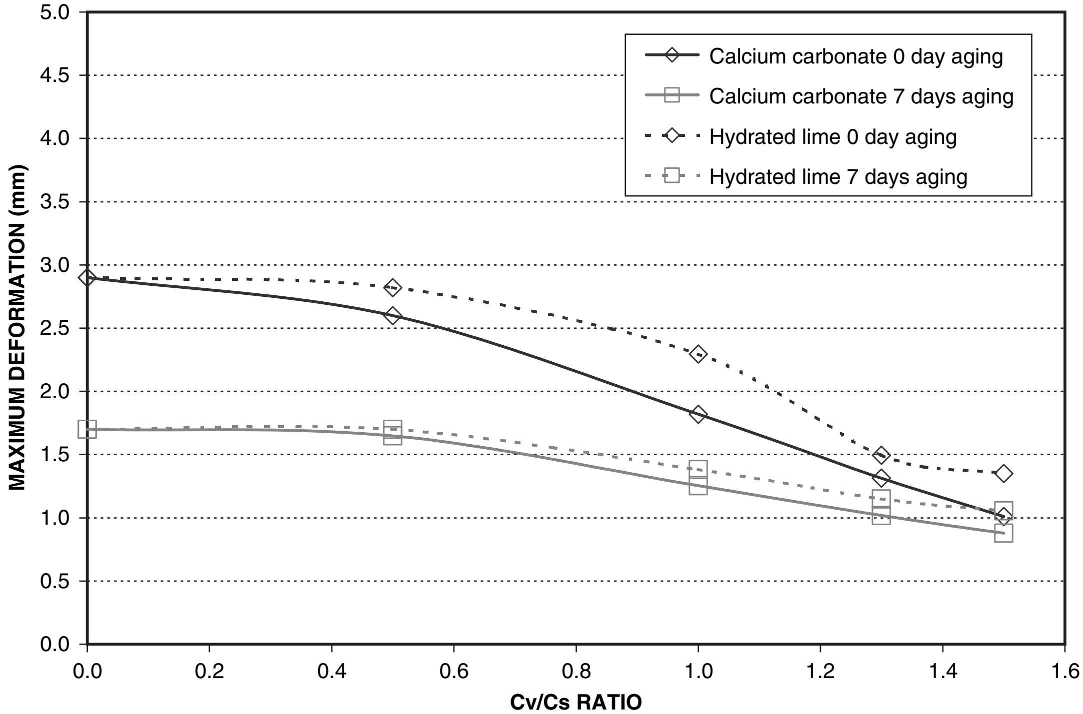

FIGURE 9 Variation of maximum deformation with volumetric concentration of filler for 0 and 7 days of aging.

means a reduction of 20% to 30%. Therefore, the optimum concentration to add to the mixture should be 20% or 30% less than the optimum defined by the nonaged curve.

For the hydrated lime, the mixture that has not been aged develops the maximum energy at the same concentration as the critical concentration ($C_v/C_s = 1.0$). This value coincides with the ideal one recommended in some specifications for the design of asphalt mixtures for this kind of filler (*6*). However, the ideal ratio for the mixture that has not been aged would be below this one (it is difficult in this case to give a specific value, although a reduction of 20% or 30%, as proposed in the preceding paragraph, seems adequate). The energy developed by the mixture with lime seems to be somewhat lower than that of the mixture with calcium carbonate, but the fact that the mixtures cannot be compared directly—as their porosities are not the same—must be taken into account.

CONCLUSIONS

All the tests performed for this paper have shown the protective effect of the fillers used (calcium carbonate and hydrated lime) against the aging of the bitumen. They have also shown the need to add the fillers to the mixture in volumetric concentrations, not by weight. Thus, their incorporation in increasing concentrations indicates a smaller decrease in the penetration, a smaller increase in the softening point, and a smaller increase in the viscosity of the bitumen, indicators that aging is less.

Nevertheless, one of the properties that could be affected by mastic aging is the cracking resistance, which is generally ignored in mixture design and characterization. Therefore, a new direct tensile test was developed by the Road Research Laboratory of the Technical University of Catalonia. This test can demonstrate the protective

TABLE 2 Characteristics and Contents of Filler Used in Study

Filler Type	Specific Gravity (g/cm^3)	Critical Concentration	Cv/Cs Ratio	Filler Content (g)	Filler/Bitumen Ratio (in weight)
Hydrated lime	2.351	0.165	0.5	5.1	0.21
			1.0	11.2	0.46
			1.3	15.2	0.63
			1.5	18.7	0.77
Calcium carbonate	2.771	0.370	0.5	14.8	0.61
			1.0	35.5	1.46
			1.3	58.0	2.39
			1.5	80.0	3.29

TABLE 3 Air Voids Contents and Densities of Mixtures

Filler Type	Cv/Cs Ratio	Air Voids Content (%)	Density (g/cm^3)
Without filler	0	27.9	1.776
Hydrated lime	0.5	27.2	1.815
	1.0	26.4	1.834
	1.3	25.9	1.844
	1.5	26.0	1.842
Calcium carbonate	0.5	25.9	1.832
	1.0	22.6	1.921
	1.3	21.1	1.964
	1.5	20.9	1.976

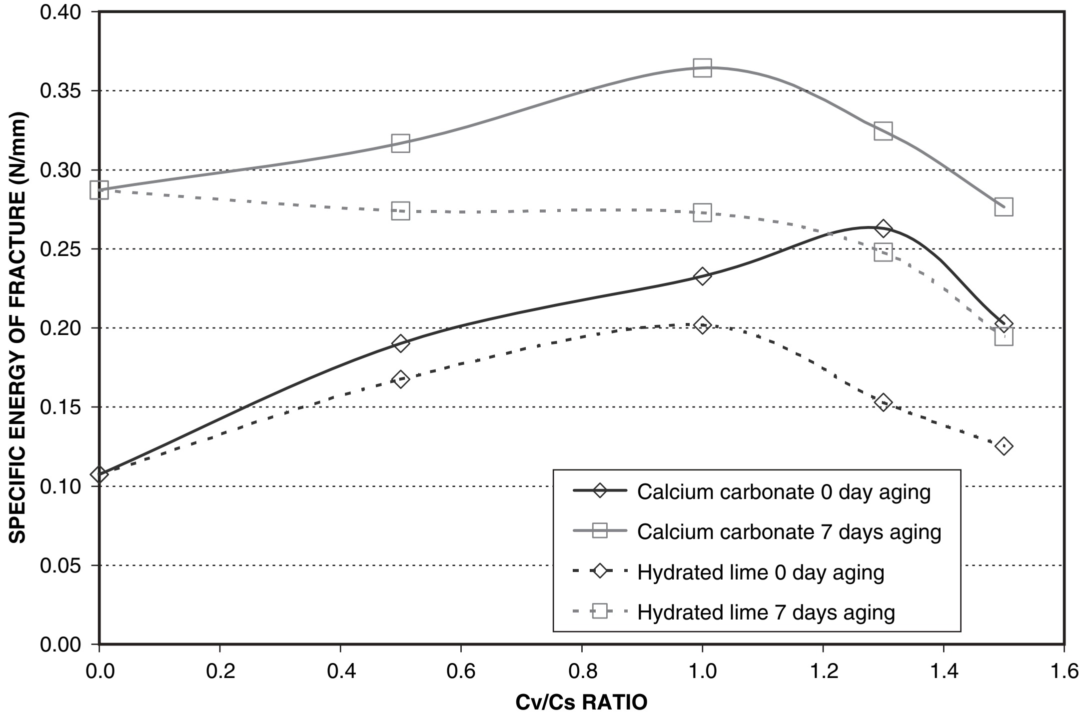

FIGURE 10 Variation of specific energy of fracture with volumetric concentration of filler for 0 and 7 days of aging.

effect of the filler. It has been observed that the hydrated lime tends to stiffen the mixture less and make it less brittle than the calcium carbonate does under the same aging conditions. This situation indicates that the lime has more protective effect than the calcium carbonate.

However, the most important conclusion resulting from the study is that the specific energy of fracture, which was assessed with the BTD test, allows us to establish a criterion for selecting the ideal content of filler volume to be added to minimize aging of the bitumen. Despite the protective role of the filler, the content must be about 20% or 30% less than the content recommended under conditions when there is no aging, so that when the mixture ages the mastic is able to build up the maximum energy possible.

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